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Dear Sir:

Enclosed please find the Final Technical Report for ONR Grant N00014-92-J-1344.



Sincerely,

James M. Valles Jr

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Assistant Professor of Physics

JMV/nyc

Encl:
cc: Ms. June A. Hawley
Dr. Donald H. Liebenberg
Administrative Grants Officer
Director, Naval Research Laboratory

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# FINAL TECHNICAL REPORT

**ONR Grant N00014-92-J-1344**

**Principal Investigator: James M. Valles, Jr.**

**Work supported by this grant has resulted in five manuscripts. These manuscripts describe in detail the technical progress made. They are:**

1. **"Electron Tunneling Determination of the Order-Parameter Amplitude at the Superconductor-Insulator Transition in 2D," by J.M. Valles, Jr., R.C. Dynes, and J.P. Garno, *Phys. Rev. Lett.* 69, 3567 (1992).**
2. **"Perpendicular Upper Critical Field of Granular Pb Films Near the Superconductor-to-Insulator Transition," by Shih-Ying Hsu and J.M. Valles, Jr., *Phys. Rev.* B47, 334 (1993).**
3. **"Magnetic-Field-Induced Pair-Breaking Effects in Granular Pb Films Near the Superconductor-to-Insulator Transition," by Shih- Ying Hsu and J.M. Valles, Jr., *Phys. Rev.* B48, 4164 (1993).**
4. **"The Proximity Effect in Ultrathin Granular Pb Films," by Shih-Ying Hsu, J.M. Valles, Jr., P.W. Adams, and R.C. Dynes, to be published in the Proceedings of LT20.**
5. **"The Coulomb Anomaly in Strongly Disordered Films," Shih-Ying Hsu and J.M. Valles, Jr., to be published in the Proceedings of LT20.**

# Electron Tunneling Determination of the Order-Parameter Amplitude at the Superconductor-Insulator Transition in 2D

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We have investigated the behavior of the superconducting energy gap $\Delta$ in ultrathin films of quench condensed Bi near the superconductor-insulator (SI) transition. From electron tunneling measurements on these films, we conclude that $\Delta$ becomes very small and approaches zero at the SI transition. We studied high-sheet-resistance films with $T_{c0}$'s as low as 0.19 K. This is a factor of 40 lower than the low-sheet-resistance film $T_{c0}$ of 6.4 K.

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As the amount of disorder is increased in a metallic system the conduction-electron states begin to localize, and the strength of the effective electron-electron interactions increases. These two effects hinder the formation of the normally superconducting state in a disordered material. The localization of the electrons opposes the formation of a coherent state over distances longer than the localization length. The enhanced repulsive electron-electron interactions will reduce the net attractive interaction that is required for Cooper pairing. In two-dimensional systems the effects of increasing disorder on the superconducting state are dramatic and eventually drive a transition from a superconducting to an insulating state [1-6].

To qualitatively describe this transition it is helpful to consider the superconducting order parameter $\Psi \propto \Delta_0^{1/2} e^{i\phi}$ where the amplitude of the order parameter, $\Delta_0$, is the zero-temperature energy gap and $\phi$ is its phase. If a film is composed of islands that are large enough to have independent superconducting properties then the superconducting state can be weakened by reducing the (Josephson) coupling between the islands. This is accomplished by reducing the tunneling probability between the islands which results in an increase in the normal-state sheet resistance, $R_\square$, of the film. Fluctuations in the phase of the order parameter ensue, which when strong enough destroy the long-range phase coherence across the film [2,6,7]. The situation has been shown to be different for the case where the morphology of the film is more homogeneous, i.e., for disorder on shorter length scales. In systems that are not too close to the superconducting to insulating transition, increases in $R_\square$ have been shown to lead to well-defined decreases in the amplitude of the order parameter [7,8] and the mean-field transition temperature, $T_{c0}$ [3-11], in direct contrast with the islanded film case. One might expect that very near the superconducting to insulating transition, fluctuations in the amplitude of the order parameter would play a dominant role in driving the transition. Recently, however, it has been argued that even in these systems, phase fluctuations dominate the physics of the superconducting to insulating transition [6,12]. In this paper we present tunneling and transport results on uniformly disordered Bi films that are very close to the superconducting to insulating transition. We find that the energy gap in these films is reduced significantly from its bulk value, showing that the amplitude of the order parameter continues to decrease at the same rate as $T_{c0}$ *even near the transition.* That is, the amplitude of the order parameter is extremely small or zero at the superconductor to insulator transition in the homogeneous case. Fluctuations in the amplitude must therefore be at least as important as fluctuations in the phase of the order parameter.

The electron tunneling and transport measurements described here were performed on films evaporated onto substrates that were held at low temperatures (4-7 K) in a cryogenic evaporator. The substrates were thermally connected to the mixing chamber of a dilution refrigerator giving a temperature range for our measurements of $0.13 < T < 7.0$ K. Contact pads and an Al strip of width 0.01 in. were deposited on the substrate prior to cooling down the apparatus. The Al strip had a small amount of Mn impurities in it to prevent it from superconducting. It was oxidized in air and served as the counterelectrode in the tunnel junctions, Al/oxide/Bi film. At low temperatures, a 1-2-monolayer film of Sb was evaporated onto the substrate and Al oxide surfaces followed by a series of Bi evaporations. The width of the Bi film was 0.1 in. so that the tunnel junction area was 0.1 in.×0.01 in. The Sb film is an insulator and helps the Bi form a very thin uniform, continuous film [13]. For example, the mass of Bi per unit area for a film with $R_\square(7\ \mathrm{K}) = 8\ \mathrm{k}\Omega/\square$ was equivalent to only four atomic layers of crystalline Bi. After each Bi evaporation the resistance of the film as a function of temperature, and the voltage dependence of the tunnel junction conductance, $G_j(V)$, were measured. The Sb film and the Al oxide serve as the tunnel barrier. The completed tunnel junction had the standard cross stripe geometry. Care was taken to assure that the measurements of $G_j(V)$ with the Bi film in the normal state were not influenced by the sheet resistance of the Bi film. With this arrangement, we can correlate all changes in the tunnel junction conductance with changes in $R_\square$ and so with changes in the film properties and not to changes in the tunnel junction barrier. We present tunneling and transport data on three Bi films, two with high $R_\square$ (near the superconducting to insulating transition), and one with a very low $R_\square$. All three were evaporated in sequence onto the same film and tunnel junction. We have obtained qualitatively similar results on Pb films in other experimental runs. In all cases, the tunnel junctions were of high quality, showing essentially no leakage current due to nontunneling conduction.

Recent measurements on ultrathin Bi films have shown that the superconducting to insulating transition occurs near $R_\square(T>T_c)\simeq 6.45$ k$\Omega/\square$ [4]. We have measured the temperature dependence of the sheet resistance and the tunneling density of states of similar uniform Bi films in order to evaluate the behavior of the amplitude of the order parameter near this superconductor to insulator transition. In Fig. 1 we plot the $R_\square(T)$ for two Bi films with $R_\square(7\text{ K})\cong 8.0$ and 5.8 k$\Omega/\square$. Both films show behavior suggesting that at $T=0$ they would be insulators ($dR/dT<0$), but then have reasonably sharp superconducting transitions. In the case of the 8.0-k$\Omega/\square$ film the minimum resistance that we measured was limited by the base temperature of our refrigerator. Clearly both films show mean-field transition temperatures that are greatly depressed from the thick Bi film value, $T_{c0}\simeq 6.4$ K, which was achieved upon further evaporation. If we take the midpoint of $R_\square(T)$ to give $T_{c0}$ then we find that $T_{c0}=0.7$ and 0.19 K for the 5.8- and 8.0-k$\Omega/\square$ films, respectively. These films are very close to the superconducting to insulating transition. In fact we find that the 8.0-k$\Omega/\square$ film *superconducts*, in contrast with earlier measurements on Bi films evaporated onto Ge [4], for which a case was made that the SI transition occurred at $h/4e^2\simeq 6.5$ k$\Omega$.

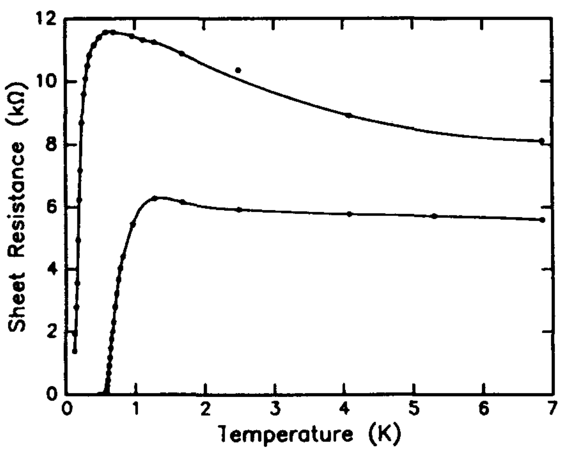


FIG. 1. Resistance as a function of temperature for two sequentially evaporated films that had sheet resistances at 7 K, $R_\square(7\text{ K})$, of 5.8 and 8.0 k$\Omega/\square$.

A slightly thinner film with higher $R_\square$ is insulating. We speculate that the discrepancy between these experiments results from the different preplating elements, Sb and Ge, that were used. Together the results support the idea that SI transition does not occur at a universal value of $R_\square$. The purpose of this paper is to study the amplitude of the superconducting order parameter on the superconducting side of the SI transition.

We can learn about the amplitude of the order parameter or the energy gap near the superconductor to insulator transition by measuring the tunneling density of states of these films. The conductance of a normal-metal/oxide/superconducting film tunnel junction can be written [2]

$$G_j(V,T)\propto N_2\int_{-\infty}^{+\infty} N_S(E)N_N(E)\frac{df(E+eV)}{dE}P(E)dE\,, \tag{1}$$

where $N_2$ is the density of states of the Al electrode which is assumed constant, $N_S(E)=|E|/(E^2-\Delta^2)^{1/2}$ is the BCS density of states, $N_N(E)$ is the normal-state density of states, $E$ is the electron energy measured from the Fermi energy $E_F$, $f$ is the Fermi function, and $P(E)$ is the tunneling probability which on the voltage scales considered here is approximately constant. At low temperatures ($T\ll T_{c0}$) the Fermi functions are sharp and Eq. (1) reduces to

$$G_j(V)\propto N_2N_S(eV)N_N(eV)\,. \tag{2}$$

*Thus, a measurement* of the voltage dependence of the conductance at low temperatures gives a measure of the density of states of the film as a function of energy.

In Fig. 2 the conductance of the tunnel junction for the 5.8- and 8.0-k$\Omega/\square$ films at $T=0.13$ K and a very low $R_\square$

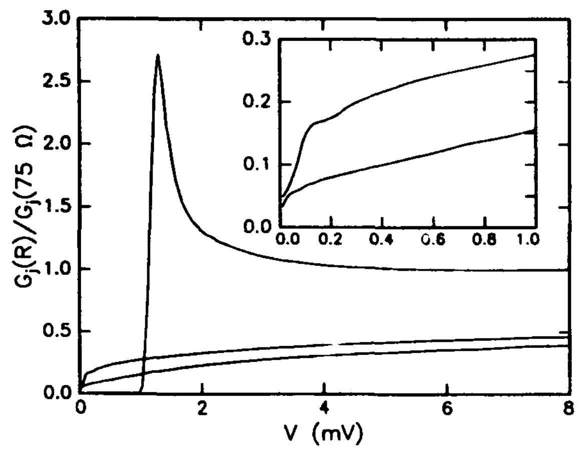


FIG. 2. Tunnel junction conductances as a function of voltage for the 8.0-k$\Omega/\square$ (lowest curve) and 5.8-k$\Omega/\square$ (middle curve) films at $T=0.13$ K and for a $R_\square(7\text{ K})=75$ $\Omega/\square$ film (upper curve) at $T\approx 0.3$ K. The junction conductances are normalized by that of the 75-$\Omega/\square$ film measured at a temperature above its $T_c$. Inset: The data for the 5.8- and 8.0-k$\Omega/\square$ films on an expanded scale.

film, 75 $\Omega/\Box$, at $T<1.0$ K, are plotted as a function of voltage. The tunnel junction conductances of the two high $R_\Box$ films have been normalized to the normal-state conductance for the very low $R_\Box$ film which is essentially voltage independent over this range and equal to 0.063 $k\Omega^{-1}$. This procedure eliminates $P$ and is legitimate because the 75-$\Omega/\Box$ film is the result of subsequent evaporations on the same junction. There are three important features of these data. First, the overall tunnel junction conductance of the high $R_\Box$ for these films is smaller over this voltage range than that of the low $R_\Box$ film, e.g., $G_j$(5 mV, 5.8 k$\Omega$)$\simeq$0.4$G_j$ (5 mV, 75 $\Omega/\Box$). Second, there is a "cusp" in $G_j$ at $V=0$. These features are characteristic of tunneling into disordered thin films and have been studied extensively [14,15]. Finally, there is a bump in $G_j(V)$ at a low voltage in the 5.8-k$\Omega/\Box$ film which appears below the transition temperature of this film. This bump is clearer in the expanded data in the inset of Fig. 2.

The first two features are qualitatively what one would expect to see based on observations made in tunneling measurements on lower $R_\Box$ films. It has been shown that in low $R_\Box$ films, $R_\Box \ll h/4e^2$, disorder-enhanced Coulomb interactions lead to logarithmic corrections to the density of states of the form

$$\frac{\delta N_n(E)}{N_0}=-\frac{1}{16\pi}\frac{R_\Box}{(6.5\,\mathrm{k}\Omega)}\times\ln\left(\frac{|E|}{\hbar D(2\pi/d)^2}\right)\ln\left(\frac{(2\pi)^2|E|}{\hbar D\kappa^4 d^2}\right) \quad (3)$$

in the weak disorder limit [15]. $D$ is the electronic diffusivity, $\kappa$ is the 2D screening wave number, and $d$ is the film thickness. These corrections lead to a cusp and an overall reduction in $G_j(V)$ similar to what we observe in this strongly disordered film. In earlier work, we showed that the overall reduction in $G_j(V)$ in junctions on films with $R_\Box$ up to 500 $\Omega/\Box$, for which the density of states correction at 5 mV (the average phonon energy relevant for superconductivity in Pb) is about 10%, agrees quantitatively with this model [9]. Thus, we believe that the data on these high $R_\Box$ films show that disorder-enhanced Coulomb interactions suppress the tunneling density of states very strongly, by more than 50% at 5 meV.

Near $V=0$ a "bump" can be observed in $G_j(V)$ for the 5.8-k$\Omega/\Box$ film at $T=0.18$ K $<T_{c0}=0.7$ K. This is more clear on the smaller voltage scale of the inset of Fig. 2. We associate this bump with the peak in the density of states at $V=\Delta/e$ in this superconducting film. In Fig. 3 we plot $G_j(V)$ at $T=0.13$ K normalized to $G_j(V)$ at 0.61 K over a smaller voltage scale. This normalization removes most of the energy dependence of the normal-state density of states and brings out the structure in the superconducting density of states. While this normalization procedure is only approximate, we believe that it is

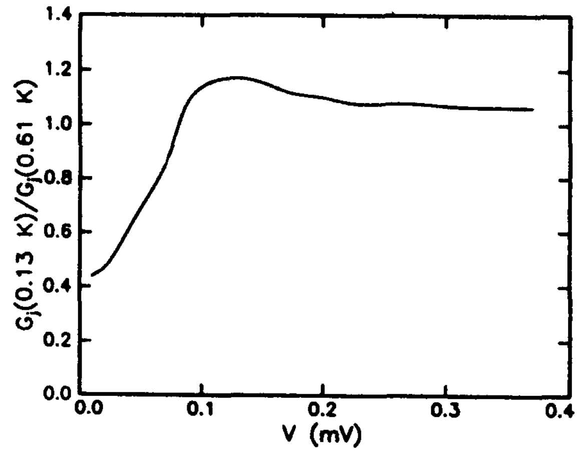


FIG. 3. Tunnel junction conductance of the 5.8-k$\Omega/\Box$ film at $T=0.13$ K normalized to its conductance at $T=0.61$ K.

sufficiently good to give us a superconducting density of states that is very near the true answer. The shape of the normalized conductance qualitatively resembles that expected for tunneling into a BCS superconductor. There is a peak in the normalized conductance and a reduction of the number of states near the Fermi energy. The deviations from perfect BCS behavior may be partially due to the normalization procedure that we have had to use and may also reflect the fact that the density of states in a strongly disordered superconducting film may be different from the BCS form because of quasiparticle lifetime or fluctuation effects [2,16,17]. For such films where $T_c$ is so severely reduced, it is not unexpected that fluctuation effects are eventually observed. Nevertheless, the signature of the superconducting energy gap is clear. We can estimate the size of the energy gap from these data by measuring the voltage at which the normalized conductance crosses 1 [18]. We get $\Delta\simeq0.082$ meV. Taken with the resistive midpoint we get $2\Delta_0/k_BT_c\simeq2.7$. In view of the substantial normalization effects this is reasonably close to the BCS value. This implies that this high-$R_\Box$ Bi film has nearly BCS-like characteristics.

On the highest $R_\Box$ film (8.0 k$\Omega/\Box$), we indeed observe the beginning of the opening of a gap as shown in the inset of Fig. 2. Because we are not at sufficiently reduced temperature, $T/T_c0$, and the potential normalization errors are more severe, however, more quantitative analysis seems inappropriate at the moment.

In earlier work, on uniform ultrathin films of Pb and Sn, the reduction in $T_c$ with $R_\Box$ was shown to be accompanied by a reduction in $\Delta$ in such a way that $2\Delta_0/k_BT_{c0}\simeq$const for $R_\Box\lesssim3$ k$\Omega/\Box$ [8,9]. The films showed BCS behavior with reduced transition temperatures. The reduction in $\Delta$ and $T_{c0}$ correlated with a reduction in the tunneling density of states due to disorder-enhanced electron-electron interactions. The size of the reduction in $N_n(E)$ has been shown to be sufficient to account for the depression of $T_c$ and $\Delta$ in these films [9,19]. Our results here suggest that this trend continues up to the superconducting to insulating transition. That is, $T_{c0}$ and $\Delta_0$ decrease at the same rate even in films near the superconducting to insulating transition and at the transition, $T_{c0}$ and $\Delta$ go to zero. The concomitant reduction in the tunneling density of states at the average electron-phonon frequency implies that the reduction in $T_{c0}$ and $\Delta_0$ are due to disorder-enhanced electron-electron interactions [9,19]. We conclude that the amplitude of the order parameter is extremely small or zero at the superconducting to insulating transition in these homogeneous films. We add, however, that in the granular case we reach a very different conclusion. The amplitude $\Delta_0$ is finite at the transition and $T_{c0}$ is not well defined [2,7].

Recently, theories of the SI transition have been proposed that argue that the transition is driven by increased phase fluctuations of the order parameter [12]. In this picture, below a critical resistance but in the critical regime the Cooper pairs form a Bose superfluid. Above the critical resistance these bosons localize and a superfluid of vortices exists. Since the vortices move freely through the system there are large phase fluctuations, dissipation, and, consequently, insulating behavior. What is essential to this picture is that the amplitude of the order parameter must be finite and robust in the vicinity of this transition. The formation of vortices requires a well-developed order-parameter amplitude. Our work here shows that in uniform Bi films, the amplitude of the order parameter (energy gap) goes to zero or becomes extremely small near the transition, i.e., it is not well developed. It is small enough that fluctuations in *its* amplitude should become significant. While we cannot rule out from the data of Fig. 3 that there may be some fluctuation effects observed, it is clear that $\Delta \rightarrow 0$ as $T_c \rightarrow 0$ and there are fermions in the system close to the transition. Thus, we argue that a theoretical description of the superconducting to insulating transition in these uniform Bi films must take fluctuations in the amplitude as well as the phase of the order parameter into account. We emphasize that these tunneling measurements show very clearly that in this uniform case $\Delta \rightarrow 0$ at the transition, unlike in the granular case where it has been shown that $\Delta$ remains finite through the transition [2].

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# Perpendicular upper critical field of granular Pb films near the superconductor-to-insulator transition

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We have measured the perpendicular upper critical field, $H_{c2}(T)$, in ultrathin granular Pb films with normal-state sheet resistances, $R_N$, that range from 10 Ω to 11 kΩ. The films with the highest $R_N$ are near the threshold for the superconductor-to-insulator ($S$-$I$) transition. We compare our results to calculations for granular superconductors and obtain values for the grain size and coherence length characteristic of a single grain. In addition, we present a measurement of the ratio of the intergrain Josephson-coupling energy to the single-grain condensation energy near the $S$-$I$ transition in these films. The small value of this ratio, 0.05, at $R_N=11$ kΩ is consistent with the picture that fluctuations in the phase of the order parameter dominate the $S$-$I$ transition in these films.

Much recent work has shown that the introduction of disorder into ultrathin superconducting films weakens and destroys their superconducting properties and renders them insulating at $T=0$.[1-7] The qualitative nature of this superconductor-to-insulator ($S$-$I$) transition depends strongly on the morphology of the particular films under study. In films composed of grains that are weakly coupled to one another but individually large enough to support bulk superconducting order-parameter amplitudes, the $S$-$I$ transition occurs when the phases of the order parameters of the individual grains become uncorrelated.[1-3,8,9] The amplitude of the order parameter remains finite[1] and phase fluctuations drive the transition. In more homogeneously disordered superconductors, the amplitude of the superconducting order parameter decreases with increasing normal-state sheet resistance, $R_N$.[4,5] For high enough $R_N$, the amplitude of the order parameter disappears[10] and the mean-field transition temperature, $T_{c0}$, goes to zero.[5-7] Fluctuations in the amplitude of the order parameter are important in driving this $S$-$I$ transition. Unfortunately, not all films fall neatly into one of these two categories. Measurements that can reveal information about the structure of superconducting films near the $S$-$I$ transition, therefore, are important.

Earlier work on three-dimensional (3D) granular superconductors has shown that measurements of the upper critical field, $H_{c2}$, provide direct information about their structure.[11-14] In a dirty type-II superconductor:

$$H_{c2}(T)=\frac{\Phi_0}{2\pi\xi(T)^2}\ , \tag{1}$$

where $\Phi_0$ is the flux quantum and $\xi$ is the Ginzburg-Landau coherence length. Physically, it is the distance that an electron diffuses during the Ginzburg-Landau relaxation time.[12] If there is macroscopic structure in a superconductor that limits the diffusion of an electron during this time then it will affect $\xi$ and, hence, $H_{c2}(T)$.

With this in mind, we have measured the perpendicular upper critical field, $H_{c2}(T)$, in granular Pb films to learn about their structure. We find that at low $R_N$, $H_{c2}$ depends linearly on $T$ over a relatively large temperature range as expected for a dirty type-II superconductor that is homogeneously disordered. With increasing $R_N$, the size of the linear region shrinks and negative curvature develops at lower temperatures. The strength of the negative curvature increases toward, but does not reach, the square-root temperature dependence expected for perfectly isolated grains. Nevertheless, the increasing curvature reflects the presence of granular structure in these films. From these data we obtain a measurement of the radius of the smallest grains in the system, the coherence length on a grain, and a measurement of the ratio $E_j/E_{\rm con}$, the relative strength of the intergrain Josephson-coupling energy, and the single-grain condensation energy. At $R_N=11$ kΩ, this ratio is $\approx 0.05$, a small value that is consistent with the picture that the $S$-$I$ transition in granular Pb films is driven primarily by phase fluctuations.

Granular Pb films with small grain sizes were desired for these experiments. To achieve this, the Pb films were fabricated by thermal evaporation onto glass substrates that were held at a temperature, $T$, near 8 K. The substrates were heat sunk to a helium pot that sits in a vacuum can that is centered in the bore of a superconducting magnet and immersed in liquid helium. The cryopumping action by the surrounding can walls reduces the pressure of gases other than helium to $<10^{-10}$ torr. Hence, the films are free of contaminants such as $O_2$. These films became electrically continuous when the mass per unit area of Pb deposited, as measured *in situ* with a quartz microbalance, was equivalent to a thickness $d\approx 60$ Å of bulk crystalline material. Similar films have been used in numerous studies of the $S$-$I$ transition.[1-3]

Four terminal measurements of the sheet resistance of the Pb films were performed *in situ* using standard dc and ac lock-in techniques. In the following, we present data obtained in two separate experiments. A given experiment consists of a series of evaporations of Pb to form a single film with transport measurements performed between evaporations.

The normal-state sheet resistance of the films measured at 8 K, $R_N$, depends exponentially on $d$ for $R_N > 300\Omega$. In this regime, the film is composed of islands and $R_N$ is dominated by interisland electron tunneling rates which depend exponentially on the distances between islands.[1,14] The resistivity of the individual islands contributes very little to $R_N$. At lower sheet resistances, $R_N$ depends much less strongly on $d$, indicating that interisland tunneling processes do not dominate $R_N$ and the resistivities of the individual islands become important.

Typical superconducting transitions for films with $R_N = 230\Omega$ and 10 k$\Omega$ in 0- and 2-T magnetic fields applied perpendicular to their planes are shown in Fig. 1. In both cases, a drop in resistance from $0.9R_N$ to $0.1R_N$ occurs over a relatively narrow temperature range. This range increases with $R_N$ and reaches 0.5 K at $R_N = 11$ k$\Omega$. We associate this drop with the mean-field transition of the superconducting films. We designate $H_{c2}$ as the field at which the sheet resistance has dropped to $R_N/2$. Choosing a different criterion, such as $R_N/10$, has very little impact on the results that we present here. Below $0.1R_N$, there is a resistive tail that increases in size with increasing $R_N$. Eventually, this tail dominates the transition and makes the determination of the mean-field transition temperature ambiguous. For this reason, we only present data on films with $R_N \leq 11$ k$\Omega$.

In Fig. 2, we present our measurements of $H_{c2}$ for films with $10\Omega \leq R_N \leq 11$ k$\Omega$. We have plotted $H_{c2}$ as a function of $T/T_{c0}$ since the zero-field transition temperature $T_{c0}$ in these films depends on sheet resistance. There are three features of these data on which we will concentrate. First, near $T_{c0}$, except perhaps in the three highest $R_N$ films, $H_{c2}(T)$ depends linearly on temperature. Second, $H_{c2}(T)$ develops curvature farther below $T_{c0}$ and this curvature increases with increasing $R_N$. Finally, $H_{c2}$ increases with $R_N$ and appears to saturate at the higher $R_N$ at low temperatures. This behavior closely resembles that observed in three-dimensional granular systems as a function of resistivity.[11,14] In this paper, we use the results of these earlier studies to obtain a picture of the structure of two-dimensional films that are near the $S$-$I$ transition.

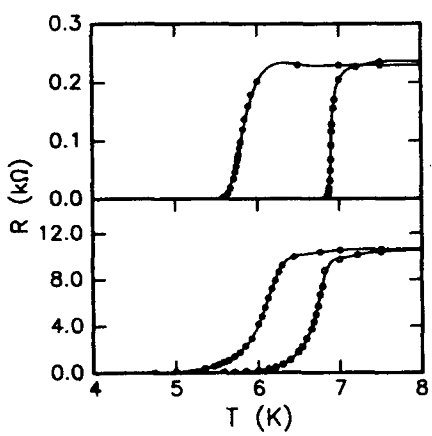


FIG. 1. Sheet resistance as a function of temperature for two Pb films with different $R_N$. The open and solid circles were obtained from 0- and 2-T magnetic fields applied perpendicular to the plane of the films. The lines are guides.

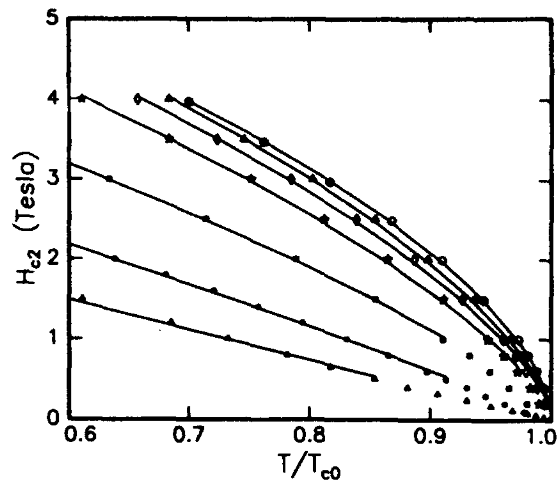


FIG. 2. Perpendicular upper critical field as a function of temperature for films with different $R_N$. The values of $R_N$, $T_{c0}$ are the following: open circles, 11.3 k$\Omega$, 6.67 K; open triangles, 1.25 k$\Omega$, 6.79 K; diamonds, 572$\Omega$, 6.83 K; stars, 292$\Omega$, 6.88 K; squares, 100$\Omega$, 6.96 K; solid circles, 34$\Omega$, 7.05 K; solid triangles, 10$\Omega$, 7.05 K.

To qualitatively understand our data we appeal to models of ordered Josephson junction arrays of spherical grains.[11] These models have been successful in describing $H_{c2}(T)$ in granular 3D superconductors in weak magnetic fields and strong magnetic fields with weak intergrain coupling. For the former case, the granular films should behave like dirty type-II superconductors for which $H_{c2}(T)$ is linear in temperature with[11,13,14]

$$\xi(T) = 0.855(\xi_{cl} l)^{1/2}\left[1 - \frac{T}{T_{c0}}\right]^{-1/2}, \tag{2}$$

where $l$ is the mean free path, $\xi_{cl}$ is the zero-temperature clean limit coherence length, and $H_{c2}(T)$ is given by Eq. (1). This formula applies when the length scale characterizing the disorder in a type-II superconductor (e.g., the average grain radius in a granular film) is shorter than $\xi(T)$, the effective coherence length for the film. This will always be satisfied sufficiently close to $T_{c0}$ where $\xi(T)$ diverges. Physically, the upper critical field is the field at which the cores of the field-induced vortices overlap and the amplitude of the superconducting order parameter is depressed to zero throughout the sample.

We fit the linear region of $H_{c2}(T)$ for each of the four lowest $R_N$ films and plotted the quantity $|T_{c0}dH_{c2}/dT|^{-1}$ versus $k_F l$ in Fig. 3 to check whether the data agree with Eq. (2). $l$ was determined using the Drude model for the resistivity and the free-electron value for $k_F$ for Pb. In addition, we have included approximations to the slopes of $H_{c2}$ at $T_{c0}$ for the highest $R_N$ films. These points are only approximate lower bounds for $|T_{c0}dH_{c2}/dT|$, as the linearity of $H_{c2}$ could not be established for these films. For all of the data, $\xi(0)$ ranges from approximately 16 to 100 Å. The solid line represents the expected behavior for a superconductor with $\xi_{cl} = 640$ Å. The data asymp-

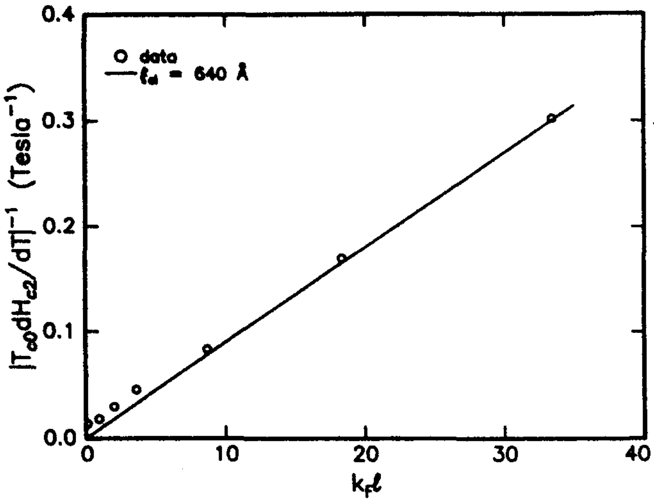


FIG. 3. Slope of $|T_{c0}(dH_{c2}(T)/dT)|^{-1}$ vs $k_F l$. The line has been calculated for an ideal dirty type-II superconductor with $\xi_{cl}=640$ Å.

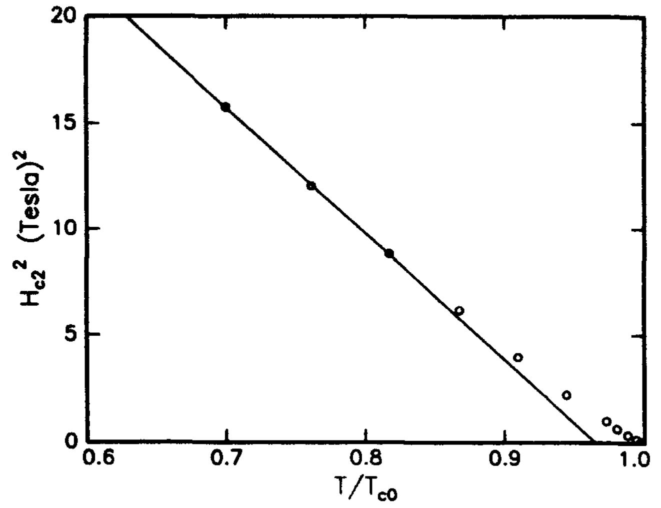


FIG. 4. $H_{c2}^2$ vs $T/T_{c0}$ for the 11-kΩ film. The line has been fit to the three highest field points to estimate the asymptotic behavior.

totically approach this line with increasing $l$. This agreement with Eq. (2) seems very reasonable. The deviations from the straight line occur at the shortest mean free paths where the determination of $|T_{c0}dH_{c2}/dT|$ is the most uncertain. For bulk Pb, $\xi_{cl}=830$ Å, which is near the measured value. The slight difference may be the result of the stronger-coupling behavior of quench condensed Pb films compared to that of crystalline bulk Pb.

Farther below $T_{c0}$, $H_{c2}(T)$ deviates from simple type-II behavior showing negative curvature that increases with $R_N$. This is qualitatively consistent with what should occur in the limit of weak intergrain coupling and strong magnetic fields where

$$H_{c2}(T)^2=\frac{5}{3}\left[\frac{\Phi_0}{2\pi\xi_{gr}R}\right]^2\left[1-\frac{2\xi_{gr}^2}{t^2}\right], \quad (3)$$

where $t$ is the intergrain coupling length, $\xi_{gr}$ is the coherence length of an individual grain, and $R$ is the radius of a spherical grain in the film.[11] For very weak coupling, $t$ diverges and $H_{c2}(T)$ assumes the value and square-root temperature dependence which is appropriate for an isolated grain.[11,13,14] The ratio $2\xi_{gr}^2/t^2$ therefore gives a measure of the degree to which the grains of the film are isolated from one another. At $T=0$, this ratio can be shown to be approximately equal to $(0.7)E_j/E_{con}$, where $E_j$ is the intergrain Josephson-coupling energy and $E_{con}$ is the condensation energy for a single grain in a square lattice. The physical picture of the upper critical field in this limit is different from the strong-coupling limit. This is because the cores of the field-induced vortices primarily reside in the spaces between grains and not on them. In this case, an applied field uniformly depresses the order-parameter amplitude on the grains throughout the sample. Below $H_{c2}$ there are no "normal vortex core" regions on the grains.

We have plotted $H_{c2}^2$ versus $T/T_{c0}$ for the 11-kΩ film in Fig. 4 in order to compare our data to Eq. (3). For agreement, the data should approach linear behavior at high fields and low temperatures. The highest $R_N$ film satisfies these criteria. The lower $R_N$ films do not show this behavior as clearly. The solid line in Fig. 4 represents the best fit to the asymptotic high-field form for this film.

The parameters obtained from the fit to the 11-kΩ film provide us with important quantitative insight into the properties of this system near the $S$-$I$ transition. The $y$-axis intercept of the linear fit to these data gives $H_{c2}(0)=7.55$ T and the slope of the line gives the zero-temperature upper critical field for an isolated single grain in the film, $H_{iso}(0)=7.68$ T. $H_{iso}(0)>H_{c2}(0)$ as it should be according to Eq. (3) and the difference in these two fields should be related to the degree to which the individual grains in the film are coupled to one another. To show this, we rewrite Eq. (3) at $T=0$:

$$\frac{H_{iso}^2-H_{c2}^2}{H_{iso}^2}=0.7\frac{E_j}{E_{con}}, \quad (4)$$

where $H_{iso}^2=5\Phi_0^2/3(2\pi\xi_{gr}R)^2$. Thus, the data imply for the 11-kΩ film that $E_j(0)/E_{con}(0)\approx 0.05$. To our knowledge, this is the first measurement of this ratio in a disordered thin-film superconductor near the $S$-$I$ transition. We discuss its importance in more detail below. Also, using standard expressions, $E_j=\pi\hbar\Delta_0/4e^2R_N$,[10] and $E_{con}=V_g N(E_F)\Delta_0^2/2$, where $\Delta_0=1.4$ meV is the Pb zero-temperature energy gap, $V_g=4\pi R^3/3$ is the volume of a grain, and $N(E_F)$ is the density of states at the Fermi energy in Pb we can estimate $R$ and $\xi_{gr}$. At $R_N=11$ kΩ, $E_j(0)=0.41$ meV so that $E_{con}(0)=8.2$ meV. Using $N(E_F)=2.11\times10^{22}/\text{cm}^3$ eV, we get $R\approx 46$ Å. Taken with the mass thickness at which these films first become electrically continuous, 60 Å, this value for $R$ implies that the grains of the film are more ellipsoidal than spherical. Finally, substitution of this radius into $H_{iso}(0)$ yields $\xi_{gr}\approx 120$ Å.

The ratio $E_j/E_{con}$ at the $S$-$I$ transition reveals a great deal about the driving force behind the $S$-$I$ transition in a particular system. It gives a relative measure of the energies required for fluctuations of the phase and amplitude of the order parameter. $E_j$ gives the phase fluctuation energy and $E_{\rm con}$ gives the amplitude fluctuation energy. In the case of our Pb films,[5,6,10] $E_j/E_{\rm con}$ is very small near the $S$-$I$ transition implying that phase fluctuations will dominate their behavior near the transition. This is consistent with earlier suggestions based on zero-field transport data in similar Pb films.[1-3] Films composed of smaller grains, but with a similar sized energy gap, and $R_N$ will tend to have a larger value of $E_j/E_{\rm con}$ near the $S$-$I$ transition since $E_{\rm con} \propto V_g$. Uniform films,[5,6,10] in some sense, are those composed of smallest grains. In this morphological limit $E_j/E_{\rm con}$ should assume the largest value at the transition and amplitude fluctuations should play their greatest role.

At first glance, it is surprising that these parameters characterizing the film are so reasonable given the idealized ordered array model from which they were derived. The films must be very disordered with random Josephson couplings between grains and a distribution of grain sizes. We speculate that the reason for this is that the transport in these films near the upper critical field occurs via single grains of nearly the minimum size that are coupled together with resistances on the order of $R_N$. These grains have the highest critical fields and hence, $H_{c2}$ is determined by them.

In summary, we have measured the perpendicular upper critical field of quench condensed granular Pb films with sheet resistances that range from 10Ω to 11 kΩ. Near $T_{c0}$, the films behave like dirty type-II superconductors that are homogeneously disordered on the scale of $\xi(T)$. Farther below $T_{c0}$ and at the highest $R_N$ the underlying granular structure of these films becomes evident in $H_{c2}(T)$. From these data we obtain a measure of the radius of the smallest grains in the system and the coherence length on a grain. We also determine $E_j/E_{\rm con}$ in the 11-kΩ film and argue that its small value implies that phase fluctuations should dominate the behavior of these films near the $S$-$I$ transition.

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# Magnetic-field-induced pair-breaking effects in granular Pb films near the superconductor-to-insulator transition

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We have measured the magnetic-field dependence, $0 \leq H \leq 8$ T, of the resistive transitions $R(T)$ of ultrathin granular superconducting Pb films with normal-state sheet resistances, $R_Q \leq R_N \leq 100$ kΩ, where $R_Q = h/4e^2 \simeq 6.45$ kΩ. The data suggest that the magnetic-field-induced superconductor-to-insulator transition in these films occurs in a field regime where pair-breaking effects have significantly reduced the amplitude of the superconducting order parameter. We compare our results with those obtained on systems with uniform morphologies.

Increases in the normal-state sheet resistance $R_N$ of ultrathin superconducting films leads to enhanced Coulomb interactions between the conduction electrons. These interactions act to localize the electronic states and reduce the density of states at the Fermi energy. Both of these effects are detrimental to the formation of the superconducting state. The localization of electronic states interferes with the ability of the superconducting order parameter to become phase coherent over long distances. The reduction of the density of states reduces the amplitude of the superconducting order parameter. Numerous experiments on ultrathin films of different materials and morphologies have shown that when $R_N$ exceeds $\sim R_Q = h/4e^2 \simeq 6.45$ kΩ, these detrimental effects drive a superconductor-to-insulator (SI) transition.[1-6]

Recent experiments on systems with uniform morphology[7-9] and an ordered junction array[10] have shown that superconducting films with $R_N$ near $R_Q$ can be tuned through this transition through the application of a magnetic field. In particular, experiments on InO films of microscopically uniform morphology strongly suggest that the magnetic-field-induced vortices are responsible for the transition.[7] They are mobile and hence, cause strong fluctuations in the phase of the order parameter. These results are in agreement with the predictions of a scaling theory of a zero-temperature SI phase transition.[11,12] This theory assumes that the effects of fluctuations in the phase of the order parameter and not the amplitude are relevant to the SI transition. It is important to determine whether or not this picture holds for the field-tuned transition in materials of granular, as opposed to uniform, morphology. First, the structure of the field-induced vortices and the pinning forces that reduce their mobility depend on a film's morphology. Second, films that are composed of grains that are large enough to support bulklike superconducting order parameters are inherently more susceptible to fluctuations in the phase rather than the amplitude of the superconducting order parameter.[2,3] As a result, recent theories of the SI transition[11,12] that emphasize the role of phase fluctuations appear particularly relevant to them.

We have investigated the transport properties of granular Pb films with $R_N > R_Q$ in high magnetic fields to address this question. Our results suggest that the field-tuned SI transition in granular Pb films with $R_N \geq R_Q$ occurs in a different magnetic-field regime than that probed in recent studies of this transition.[7-10] In this regime, the motion of the field-induced vortices plays a minor role and the amplitude of the superconducting order parameter has become so small that fluctuations in it become important.

The Pb films used in these experiments were thermally evaporated onto glass substrates that were maintained at a temperature near 8 K during the deposition, i.e., quench condensed. Measurements of the sheet resistance of the films were performed *in situ*. The apparatus has been described previously.[13] In the following, we present measurements on films with $R_N$ in the range 6 kΩ $< R_N <$ 100 kΩ, where $R_N \equiv R(8\text{ K})$, in perpendicular magnetic fields, 0 T $< H <$ 8.0 T. The mass per unit area of these films was approximately equivalent to that of a 60-Å-thick film of bulk Pb.

While the metastable nature of these films has precluded any direct structural studies of them, a great deal of indirect evidence indicates that they are granular. This morphology ensures that the amplitude of the superconducting order parameter is robust in the sense that external perturbations induced by changes in $R_N$ or magnetic field should primarily affect the phase of the superconducting order parameter and not its amplitude. This is in contrast to the case of uniform films for which the amplitude of the superconducting order parameter decreases as the SI transition is approached.[14] We *expect*, therefore, that phase fluctuations will dominate the field-tuned SI transition in granular films.

In Fig. 1 we show the evolution of $R(T)$ for a series of Pb films that span the zero-field SI transition. This evolution can be qualitatively described in terms of a disordered Josephson-junction array model.[3] At low $R_N$, $R_N \ll R_Q$, the intergrain Josephson coupling energies are strong enough that the order parameters on the individual grains can lock phases and the film can superconduct. As shown in Fig. 1, the resistive transitions, $R(T)$ of the lowest $R_N$ films are relatively sharp. Increases in $R_N$ to $\simeq R_Q$ weaken the intergrain couplings and make the order parameter more susceptible to thermal and quantum phase fluctuations. This leads to broad transitions to the superconducting state. For $R_N \gtrsim 100$ kΩ, the average in-

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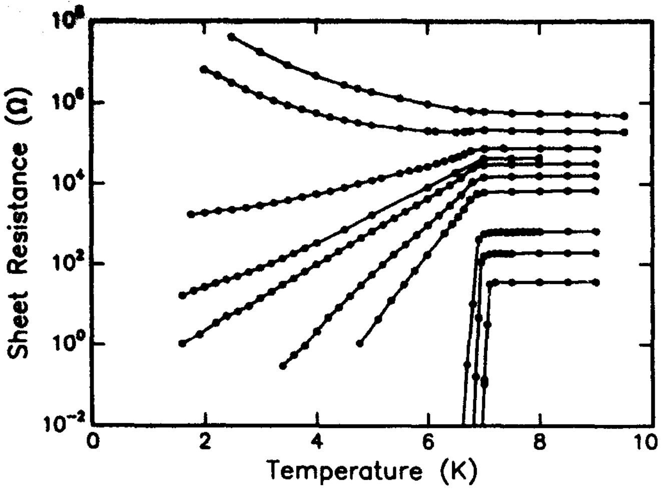


FIG. 1. Sheet resistance vs temperature for a series of Pb films evaporated in two experimental runs. Note the gradual broadening of $R(T)$ with increasing $R_N = R(8\text{ K})$ that eventually leads to insulating behavior.

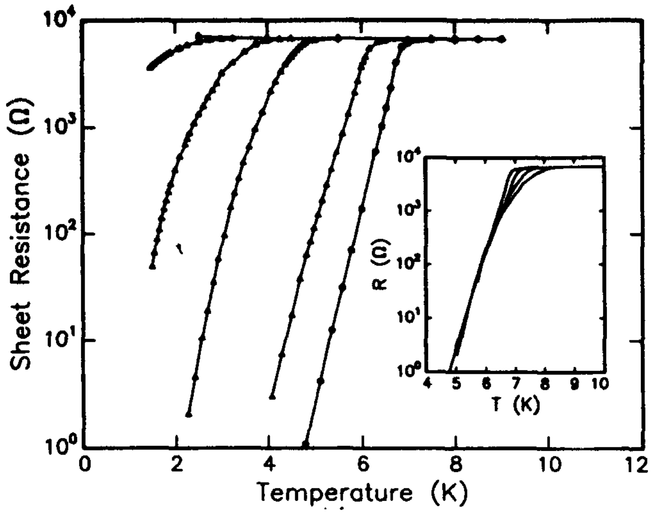


FIG. 3. Sheet resistance as a function of temperature for $R_N = 6.8$ kΩ in magnetic fields of 0, 2, 4, 5, 6, and 8 T. Inset: $R(T)$ from the main body of the figure shifted in temperature so that their low-temperature parts line up.

tergrain coupling is so weak that the disorder enhanced repulsive intergrain Coulomb interactions enable phase fluctuations that prevent the establishment of long-range phase coherence even at $T=0$. As a result, the $R(T)$ show only quasireentrant superconducting behavior and insulate at $T=0$. Eventually, for $R_N > 400$ kΩ the quasireentrant behavior disappears and $R(T)$ increases exponentially with decreasing temperature.

We show the response of films with 6.8 kΩ $< R_N <$ 100 kΩ that are on the superconducting side of the transition or show strong superconducting tendencies to an applied magnetic field in Fig. 2. As expected, the field pushes the system toward the SI transition. The data in Fig. 2, however, demonstrate that the detailed response of $R(T)$ depends on the magnitude of the applied field. "Low" fields, $H \leq 0.5$ T, make the tail of $R(T)$ grow and hence, broaden it [see Fig. 2 (left-hand side)]. Increasing the field further does not lead to additional broadening but instead to a "shift" of $R(T)$ to lower temperatures. This is shown in Fig. 2 (right-hand side) where we have plotted $R(T)$ in 0 and 2 T for two different $R_N$. We demonstrate this high field response in much more detail in Figs. 3 and 4 where we present $R(T)$ for $R_N = 6.8$ and 32 kΩ, respectively, in fields up to 8 T. In the inset of Fig. 3, we have shifted each of the curves in the main part of the figure along the temperature axis an amount, $\Delta T(H)$, so that the tails of the $R(T)$ in all fields lie atop one another.

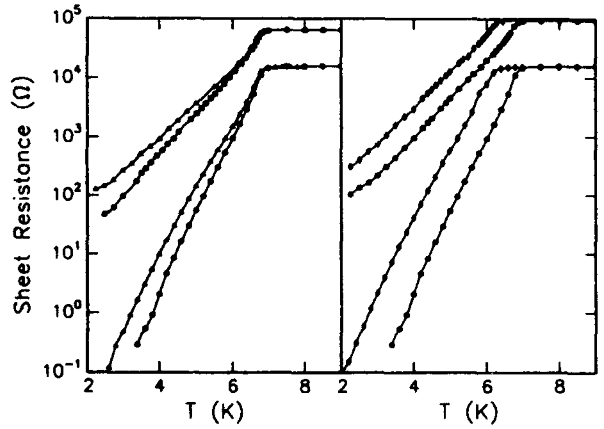


FIG. 2. Sheet resistance as a function of temperature for films with $R_N$ in the vicinity of the SI transition in magnetic field. The circles correspond to $H = 0.0$ T, the triangles on the left-hand side correspond to $H = 0.5$ T, and the diamonds on the right-hand side correspond to $H = 2.0$ T. Notice how the 0.5-T field "broadens" the 0 field $R(T)$ and how the 2.0-T field "shifts" it.

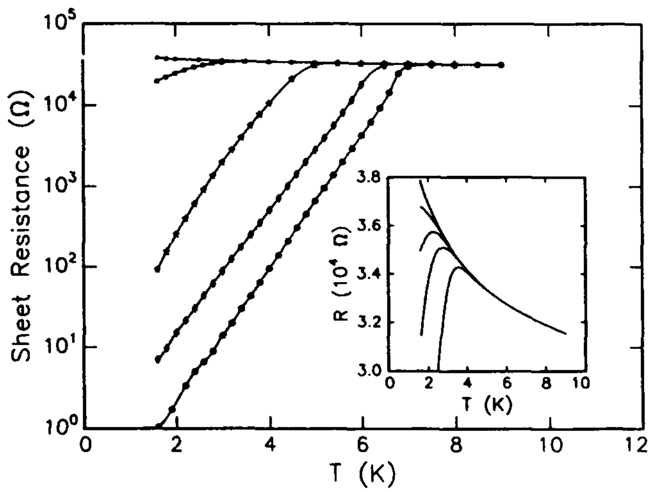


FIG. 4. Sheet resistance as a function of temperature for $R_N = 32$ kΩ in magnetic fields of 0, 2, 4, 6, and 8 T. Inset: $R(T)$ in fields of 6.0, 6.5, 6.75, 7.0, 7.5, and 8.0 T to show the approach to the normal state. The 7.5 and 8.0 T curves are nearly coincident.

Data in fields up to 4 T overlap over more than 2.5 decades of resistance.

While there are changes in the shapes of the $R(T)$ for $H \geq 4$ T, the dominant field effect still appears as a shift in the $R(T)$. Our lowest-temperature, highest-field data shown in the inset of Fig. 4 suggest that this is the case right up to $H \approx 8$ T: the field at which all signs of superconductivity in the grains of the film disappear. Qualitatively, the small changes in the shapes of the $R(T)$ that occur at these higher fields are such that they are rounder and their downward curvature is larger (see the 4 T data in Figs. 3 and 4). This latter tendency suggests that at sufficiently low temperatures, films with $R_N$ as high as 32 kΩ will superconduct in a 4-T field.

In general, the application of a magnetic field on a type-II superconductor introduces vortices that are detrimental to the superconducting state in two ways: by causing pair breaking or by causing phase slips through their motion. Pair breaking reduces the transition temperature and the amplitude of the order parameter of the superconductor. For a small pair breaking rate, $\alpha$, the resistive transition shifts by $\Delta T \simeq \pi\alpha/4$.[15] The motion of vortices in a superconductor, through flux creep or flow, increases the resistance at a given temperature in proportion to the number of vortices in the system. This usually leads to a broadening and the growth of a tail in the resistive transition that is qualitatively similar to the effect of increasing $R_N$ of a granular film (see Fig. 1) or increasing the magnetic field on InO films with $R_N \simeq R_Q$.[7] Thus, the response of $R(T)$ to low fields in granular Pb films can probably be attributed to motion of the field-induced vortices. At high fields, the pair-breaking effects of the field are more important and the field-induced vortex motion is a secondary effect.

Our earlier $H_{c2}$ studies on granular Pb films implied that the grains in films with $R_N \geq 11$ kΩ are very weakly coupled to one another.[13] In this limit, the pair-breaking effects induced by magnetic fields are dominated by the properties of the individual grains, such as their radii, and should therefore be independent of $R_N$.[16] Referring to Fig. 2 (right-hand side), it is clear that the shifts in $R(T)$ do not depend on $R_N$. From the field dependence of $\Delta T$ we have determined that the grains of the film have a lateral radius of $\simeq 46$ Å.[13]

The "normal" state that emerges as the field shifts the $R(T)$ to lower temperatures appears to be that of a disordered metal in the two-dimensional limit. We demonstrate this in Fig. 5 where we show that the *sheet conductance* of films with $R_N = 15.5$ and 32 kΩ decreases logarithmically with temperature in a field of 8 T. This temperature dependence has been observed in normal-metal films in high magnetic fields[17] and has been attributed to the effects of disorder enhanced electron-electron interactions in two dimensions.[18] These effects lead to a temperature-dependent conductance correction of the form:[18]

$$G(T)-G(T_e)=A\frac{e^2}{2\pi^2\hbar}\ln(T/T_e)\,, \qquad (1)$$

where $A$ is a constant that is less than or equal to one.[19] These electron-electron interaction effects become

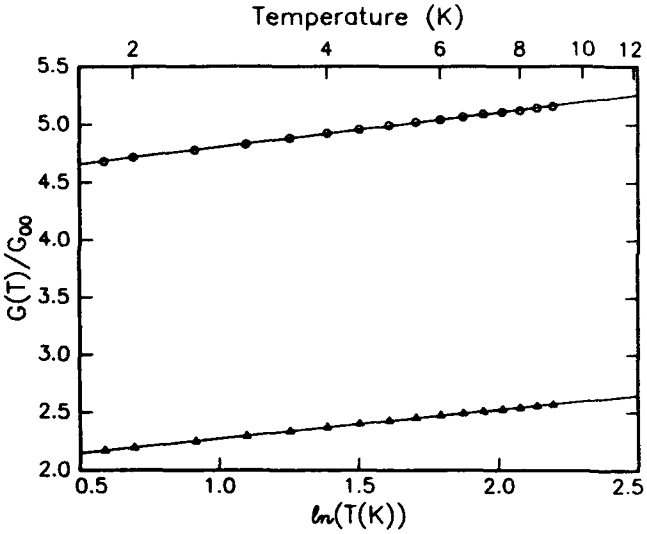


FIG. 5. Sheet conductance as a function of temperature for two films with $R_N = 15.5$ kΩ (circles) and $R_N = 32$ kΩ (triangles) in units of $G_{00} = e^2/2\pi^2\hbar$ vs the logarithm of the temperature in a field of 8 T. The solid lines are least-square fits to the data.

stronger at lower temperatures and eventually lead to the localization of the electronic states and insulating behavior. The slopes of the logarithmic fits to the data in Fig. 5 imply that $A = 0.30$ and 0.35 for the 32 and 15.5 kΩ films, respectively. Given these reasonable values, we conclude that these films have an *insulating normal state* at $T = 0$.

In this sense, these granular films can be considered superconducting insulators. Furthermore, the responses of the 6.8 and 32 kΩ films to high fields suggest that their magnetic-field-induced SI transitions are driven by the pair-breaking effects of the applied field. These pair-breaking effects reduce the amplitude of the superconducting order parameter and the $T_c$ of the individual grains of the film. Effects due to the motion of the field-induced vortices on this transition are secondary. This is in stark contrast to the behavior observed in uniform InO films[7] where the motion of the field-induced vortices is of paramount importance.

We find it surprising that the magnetic field has a more detrimental effect on the amplitude as opposed to the phase of the superconducting order parameter in granular films since increases in $R_N$ produce the opposite effect. In a similar way, the results on uniform films are counterintuitive. The InO data[7] suggest that magnetic fields induce phase fluctuations in uniform films while increases in $R_N$ reduce the order-parameter amplitude.[14] Thus, increasing disorder in or increasing the magnetic field on a film near the SI transition, in this sense, produces changes in the superconducting order parameter that are "orthogonal" to one another.

As we noted earlier, the qualitative response of $R(T)$ to magnetic fields below 2 tesla differs from that above 2 tesla. We believe that the existence of these two regimes reflects the fact that granular films have an intrinsic length scale given by the grain radius. If we model these films as Josephson junction arrays, then the minimum area of a superconducting loop will be on the same order as $\pi R^2$, where $R$ is the grain radius. This minimum loop area defines a characteristic field $H^* = \Phi_0/\pi R^2$. For $H \leq H^*$, the field response is dominated by the changes in the screening currents around loops of the array of area, $\Phi_0/H$.[16] Increases in $H$, on average, lead to increases in these currents. The flow of these currents reduces the effective Josephson coupling energies between junctions in a loop and hence, makes them more susceptible to phase fluctuations. These phase fluctuations will lead to enhanced dissipation and a broadening in $R(T)$. This is because the fractional change,

$$\delta r(H) = [R(T,H) - R(T,0)]/R(T,0) ,$$

is largest at temperatures where the loops are largest. This occurs at low temperature where the size of phase coherent regions in the film is greatest. For $H > H^*$, all loops in the film contain at least a single flux quantum and further increases in the external field do not increase, on average, the screening currents in the film. The increases in local field strength leads to increases in the pair-breaking rate dictated by the properties of the individual islands of the array.

The data in Fig. 2 show that the crossover in field response occurs near 2 T. In the context of the model above, this implies that the minimum loop in a granular Pb film has an area equivalent to a circle of radius of 180 Å. Given the grain radius of 46 Å, the minimum loop would encompass a few grains. This seems reasonable and gives us confidence that this is the physical source of the two field regimes.

The response to magnetic fields of the $R(T)$ in InO films[7] and ordered junction arrays[10] that are near the SI transition is qualitatively more similar to that observed here in the low-field regime than in the high-field regime. That is, $R(T)$ in those systems broadens in response to a field. This broadening continues through the SI transition and the $R(T)$ assume a quasireentrant shape on the insulating side of the transition. The results of these experiments agreed with recent scaling theories of the field-tuned SI transition.[11] In the case of the ordered arrays, it is clear that the experiments were performed in this low-field regime. Our experiments show that for comparable and higher $R_N$ values, disordered granular films have a field-tuned SI transition that occurs in the strong field regime. The reason for this qualitative difference between the arrays and granular films probably stems from the disorder in the latter system. In fact, recent Monte Carlo simulations predict that the critical low-temperature resistance at which the SI transition described in Ref. 11 occurs increases with disorder.[12] We speculate that the field-driven SI transition can occur in the low-field limit in granular films with $R_N \gg R_Q$ and that in this regime the transition may appear more qualitatively similar to the ordered array case. Further experimental work at lower temperatures is needed to assess this hypothesis.

In summary, we have presented measurements of the magnetoresistance of ultrathin granular Pb films with normal-state sheet resistances greater than the quantum of resistance. Our data show evidence of a magnetic-field-induced SI transition in films with $R_N$ as high as 32 kΩ. This transition occurs in a high-field regime where pair-breaking effects have significantly reduced the amplitude of the superconducting order parameter.

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[19]We can neglect weak localization effects because they are quenched in high magnetic fields.

The Proximity Effect in Ultrathin Granular Pb Films*

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**We present electron transport measurements that demonstrate that Pb films that are on the insulating side of the two dimensional superconductor to insulator transition can be made superconducting by depositing a normal metal, Ag, on them.**

## 1. INTRODUCTION

Putting a normal metal in intimate contact with a superconductor degrades some of the superconductor's properties and, under certain circumstances, enhances others. The proximity of the normal metal weakens the pairing potential and reduces the mean field *transition temperature*, $T_c$, of the superconductor[1]. On the other hand, if the superconductor has a granular morphology, then placing a normal metal among its grains can enhance the intergrain coupling strength and thereby increase the intergrain critical currents. We have investigated the effects of depositing a normal metal, Ag, onto ultrathin Pb films that are on the insulating side of the superconductor to insulator transition. The effects of the normal metal are dramatic. The insulating Pb films can be driven through the superconductor to insulator transition by the addition of Ag.

## 2. SAMPLES

The films used in these studies were evaporated onto fire polished glass substrates held at 8 K in a cryostat immersed in liquid Helium. The cryopumping action of the walls of the cryostat insures that the evaporated films are free of contaminants. Measurements of the sheet resistance as a function of temperature, $R(T)$, were performed *in situ*[2]. In what follows, we express the mass deposited in terms of the thickness of a layer of bulk material of the same areal density. For $R_N \equiv R(8K) > 300\Omega$, both $D_{Pb}$ and $D_{Ag}$, the thicknesses of the Pb and Ag layers, are smaller than the superconducting coherence length.[2]

## 3. RESULTS AND DISCUSSION

In Fig. 1a, we show logarithm of $R$ as a function of $T$ for a Pb film of 40 Å topped with increasing amounts of Ag. $R_N$ of Pb films thinner than 60 Å are too large to measure. The evolution of $R(T)$ with increasing Ag thickness is qualitatively very similar to what is observed in pure granular films.[3,4] The deposition of $\approx$ 25 Å of Ag produces the uppermost curve of Fig. 1a which rises nearly exponentially with decreasing temperature. This film was probably an insulator at $T = 0$. Subsequent evaporations of small amounts of Ag, $\approx$ 1Å, transform the film's insulating behavior into superconducting be-

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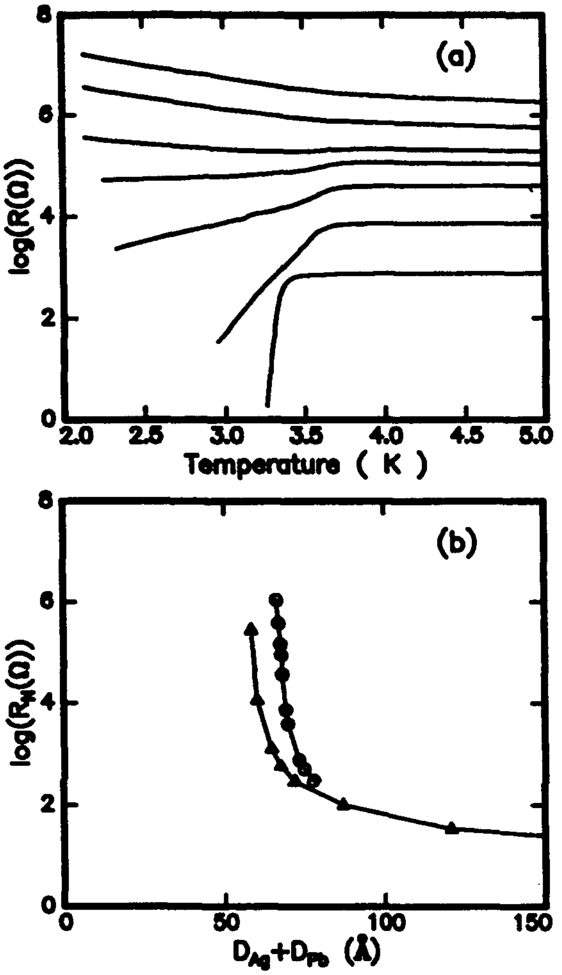


Figure 1. a)$R(T)$ for a $D_{Pb} = 40$Å film topped with different amounts of Ag. b) Film thickness dependence of $R_N$ for a pure Pb film and the films in a).

havior. The transition temperature of $\approx 3.6K$ is significantly less than the bulk Pb value of 7.2 K. Thus, these data show an insulator to superconductor transition that occurs as the amount of *normal metal* in the sample is increased.

We can learn about the morphology of this Pb/Ag system by examining the film thickness dependence of $R_N$. In Fig. 1b we plot the logarithm of $R_N$ vs. $D_{Pb} + D_{Ag}$ for pure Pb films (closed symbols) and Pb/Ag films with $D_{Pb} = 40$Å (open symbols). For the smallest thicknesses, $R_N$ decreases nearly exponentially with thickness indicating that electron tunneling processes dominate the transport and therefore, these films have a granular morphology near the insulator to superconductor transition[3]. $R_N$ would drop more slowly than exponentially if the films had a uniform morphology. Thus, the data imply that the Ag coats the Pb grains enlarging them and reducing the intergrain separation. This enhances the intergrain Josephson couplings which reduces the fluctuations in the phase of the superconducting order parameter and leads to zero resistance. The Cooper theory of the proximity effect should work this picture since it assumes that the nor and superconductor regions are in intim contact and have dimensions smaller than a coherence length. The grain $T_c$'s should be reduced by a factor of $exp(-D_{Ag}/D_{Pb}) \approx 0.5$[1] in good agreement with the data.

## 4. SUMMARY

We have used Ag, a normal metal, to transform an insulating granular Pb film into a superconductor. The deposited Ag acted to increase the intergrain Josephson coupling energies and thereby enable the superconducting order parameter to develop phase coherence over the entire sample.

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# The Coulomb Anomaly in Strongly Disordered Films*

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Electron tunneling measurements of the Coulomb anomaly in the density of states in strongly disordered quench condensed granular films are presented. The strength of this anomaly grows with increasing sheet resistance, $R_N$, at low $R_N$ but saturates at high $R_N$. We suggest that the granular morphology of these films is responsible for the saturation.

## 1. INTRODUCTION

Increases in the static disorder in metal films degrades the screening capabilities of the conduction $e^-$, increases spatial correlations among them and thus, increases the effective $e^- - e^-$ interactions. This leads to a decrease of the density of states about $E_F$. [1 − 3] In two dimensional weakly disordered systems, this decrease is given by [4]:

$$\frac{\delta N(E)}{N_0} = \lambda_\mu ln(\frac{l}{\xi}) ln(\frac{|E - E_F|\tau_{el}}{\hbar}) \quad (1)$$

where $\lambda_\mu$ depends on the strength and form of the effective $e^- - e^-$ interactions, l is the mean free path, $\xi$ is the localization length and $\tau_{el}^{-1}$ is the rate of elastic collisions. This density of states anomaly appears as an anomaly in the conductance as a function of voltage, $G_j(V)$ of tunnel junctions with a disordered film as one of the electrodes. Here, we present preliminary studies of this anomaly in films that range from the weakly disordered limit, where this theory applies, to the strongly disordered limit, where it does not. We find that film morphology plays an important role in determining the strength of this anomaly and thus, the strength of the $e^- - e^-$ interactions in strongly disordered films.

## 2. SAMPLES

The disordered films were deposited onto cooled substrates ($\sim 8K$) on which an oxidized Al strip had been previously deposited. The disordered film and Al strip served as the tunnel junction electrodes. Four terminal measurements of $G_j(V)$ were performed at 8K in situ. In fact the normal state conductance $G_j(V)$ does not depend on temperature for $eV > kT$. We have measured $G_j(V)$ of granular Pb and Sn films with $R_N$ ranging from 10Ω ($k_F l \gtrsim 1$) up to 70$k$Ω ($k_F l \sim .04$).

$G_j(V)$ can be written

$$G_j(V) = \int_{-\infty}^{\infty} N_{Al} N_{Film} \frac{\partial f(E + eV)}{\partial (eV)} P(E) dE$$

where E is the energy relative to $E_F$, f is the Fermi distribution, V is the voltage across junction, $N_{Al}$ is the density of states for the Al strip and $N_{Film}$ is the density of states for the investigated film. P(E) is the tunneling probability. We normalized all curves by the lowest $R_N$ film for each experimental run to eliminate the effects of $N_{Al}$ and P(E), so the normalized junction conductance, $G_N$, reveals corrections to $N_{Film}$ due to disorder effects.

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## 3. RESULTS

In Fig. 1 we show $G_N(V)$ vs. $\ell n(V)$ for a series of granular Sn films. The dotted lines are data and the solid lines show linear regions. These regions shrink and their slopes grow with increasing $R_N$. The functional form of $G_N(V)$ at high voltages is unclear.

We plot the slope vs. $R_N$ for Pb and Sn films in Fig. 2. At low $R_N$ these slopes grow, but at high $R_N$ they saturate. The sheet resistance at which they saturate, $R_N^*$, and the saturation value are both higher in Sn than in Pb. This saturation value appears to scale with the inverse of the film thickness.

According to Eq.(1) the slope of this $\ell n(V)$ dependence corresponds to the strength of the $e^- - e^-$ interactions and $R_N$ represents the degree of the disorder. Disorder enhances the effective $e^- - e^-$ interactions, so the slopes should grow with increasing $R_N$. We believe that this observed saturation results from a morphology change ("granular" to "uniform") that occurs near $R_N^*$. Above $R_N^*$, films are made of isolated grains and the transport properties are dominated by intergrain tunneling processes. Below $R_N^*$, the grains start to couple together and the film becomes more uniform. For the granular morphology the effective $e^- - e^-$ interactions are dominated by grain charging effects. These depend only on the intergrain capacitances which are independent of $R_N$. For the uniform morphology, where the theory applies, the effective interactions depend on $R_N$.

## 4. SUMMARY

We have measured the density of states of granular Pb and Sn films with $R_N$ up to $70k\Omega$. The strength of the coulomb anomaly saturates at high $R_N$. We attribute this effect to the granular morphology of the films.

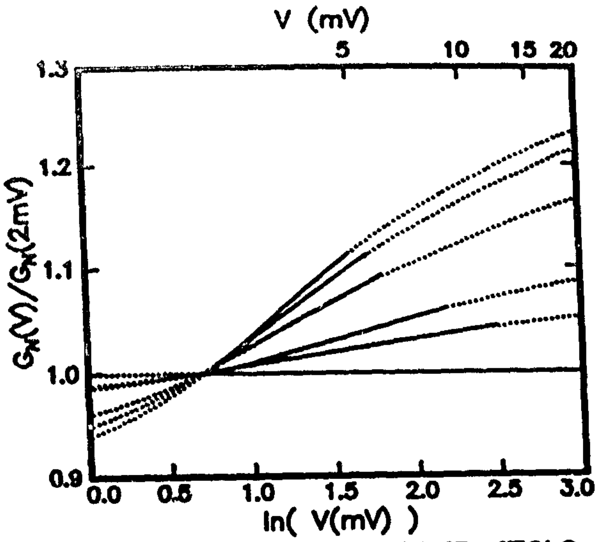


Fig. 1 granular Sn $30\Omega \le R_N \le 70k\Omega$

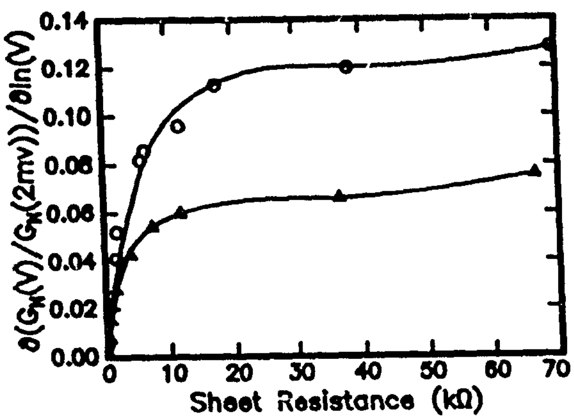


Fig. 2 O:Sn Δ:Pb

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